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Stability and Performance of Nafion-117 Membranes for the Concentration of HI/Water and HI/Water/Iodine Mixtures

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Thermochemical water splitting processes for generating hydrogen have been researched for at least thirty years in which over one-hundred chemical cycles have been proposed that use heat and/or electrochemistry to split water into hydrogen and oxygen. Proposed heat sources include nuclear reactors and solar reflectors. One of the most promising cycles is the Sulfur-Iodine (S-I) process, where aqueous HI is thermochemically decomposed into H₂ and I₂ at approximately 350 degrees Celsius. Regeneration of HI is accomplished by the Bunsen reaction (reaction of SO₂, water, and iodine to generate H₂SO₄ and HI). Furthermore, SO₂ is regenerated from the decomposition of H₂SO₄ at 850 degrees Celsius yielding the SO₂ as well as O₂. Thus, the cycle actually consists of two concurrent oxidation-reduction loops. As HI is regenerated, co-produced H₂SO₄ must be separated so that each may be decomposed. Current flowsheets employ a large amount (~83 mol% of the entire mixture) of elemental I₂ to cause the HI and the H₂SO₄ to separate into two phases. Removal of water from this system has the direct result of lowering the required quantity of I₂, thus reducing the amount of material that must be physically moved within and S-I plant. Recent efforts at the INL have concentrated on applying pervaporation through Nafion-117 membranes for the removal of water from HI/water and HI/Iodine/water feedstreams. In pervaporation, a feed is circulated at low pressure across the upstream side of the membrane, while a vacuum is applied downstream. Selected permeants sorb into the membrane, transport through it, and are vaporized from the backside. Thus, a concentration gradient is established, which provides the driving force for transport. In this work, membrane separations have been performed at temperatures as high as 132 degrees Celsius. Transmembrane fluxes of water are commercially competitive (~200 g/m²h) and separation factors ([HI]_{feed}/[HI]_{permeate}) have been measured as high as 500. All membranes studied exhibited no degradation in membrane performance during use.

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